Quantitative theory of electron-correlation effects in two-particle spectroscopies

APS

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Institut für Theoretische Physik und Astrophysik, Universität Würzburg

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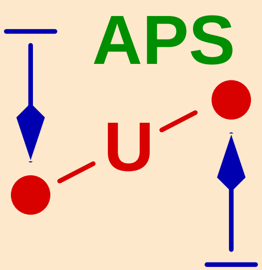

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Experiment: **M. Vonbank, K. Ertl** *MPI Garching*

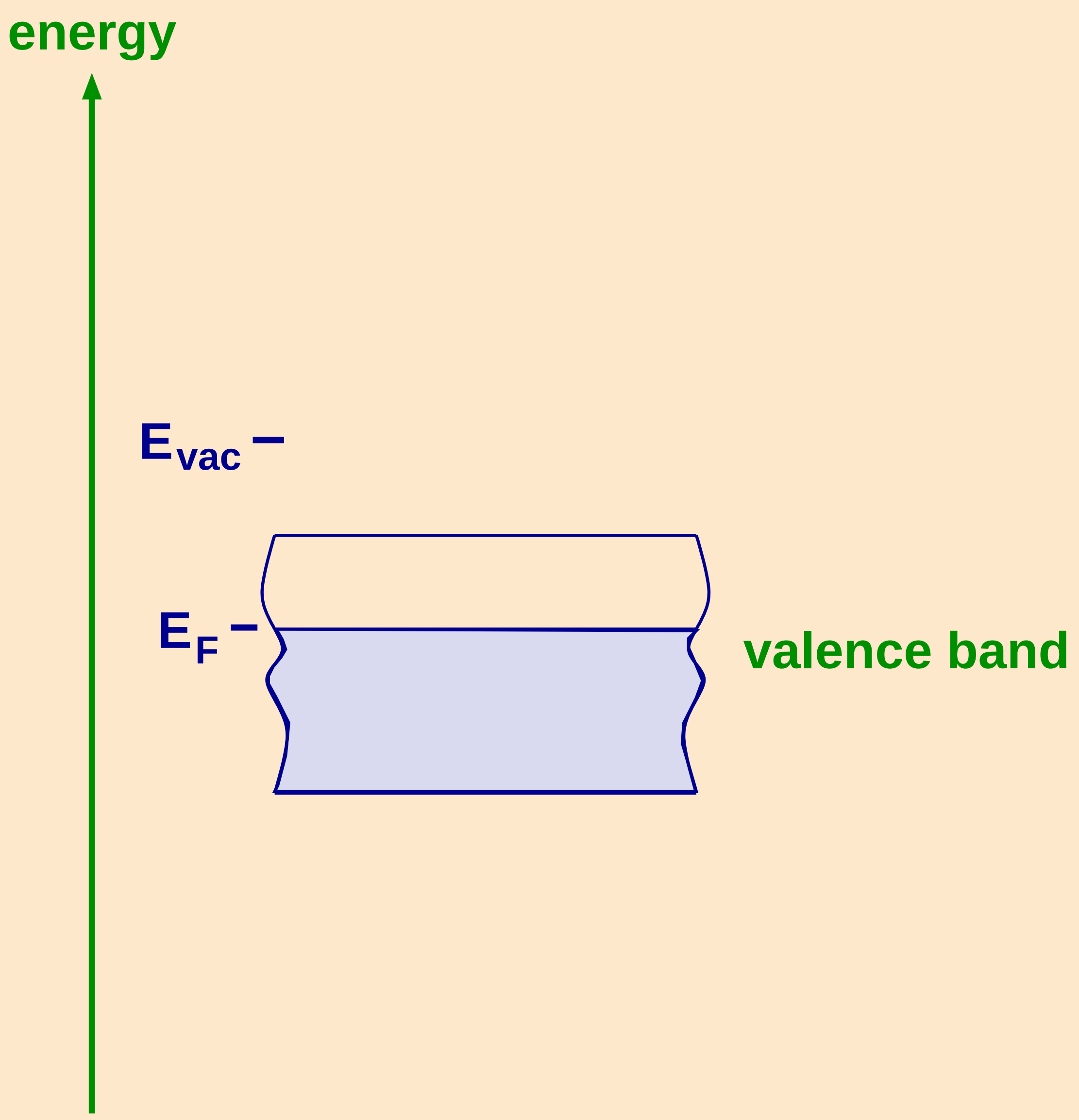
M. Donath *U Münster*

Theory: **T. Wegner, W. Nolting** *HU Berlin*

T. Schlathölter *Philips Hamburg*

J. Braun *U Münster*

motivation



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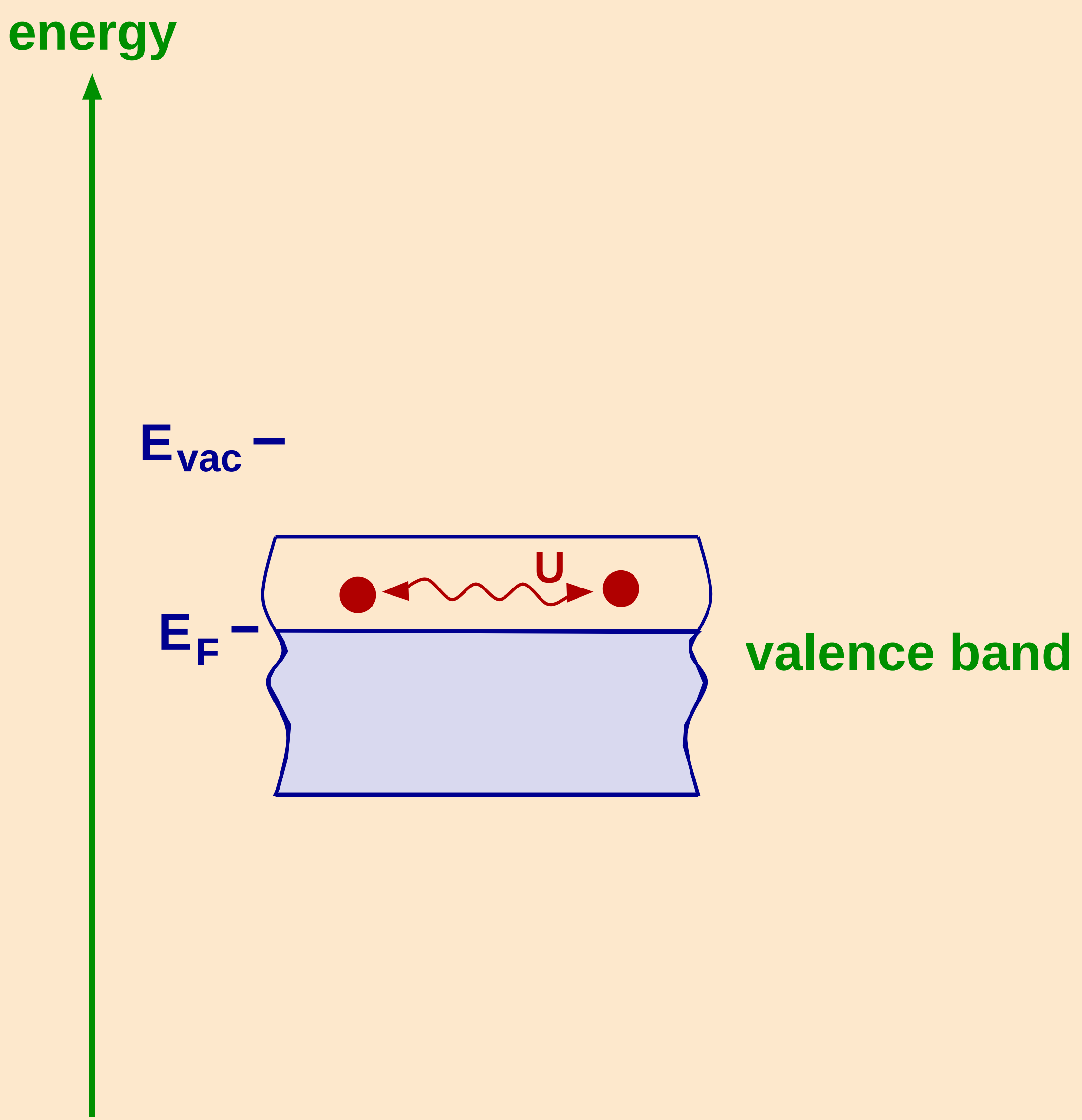

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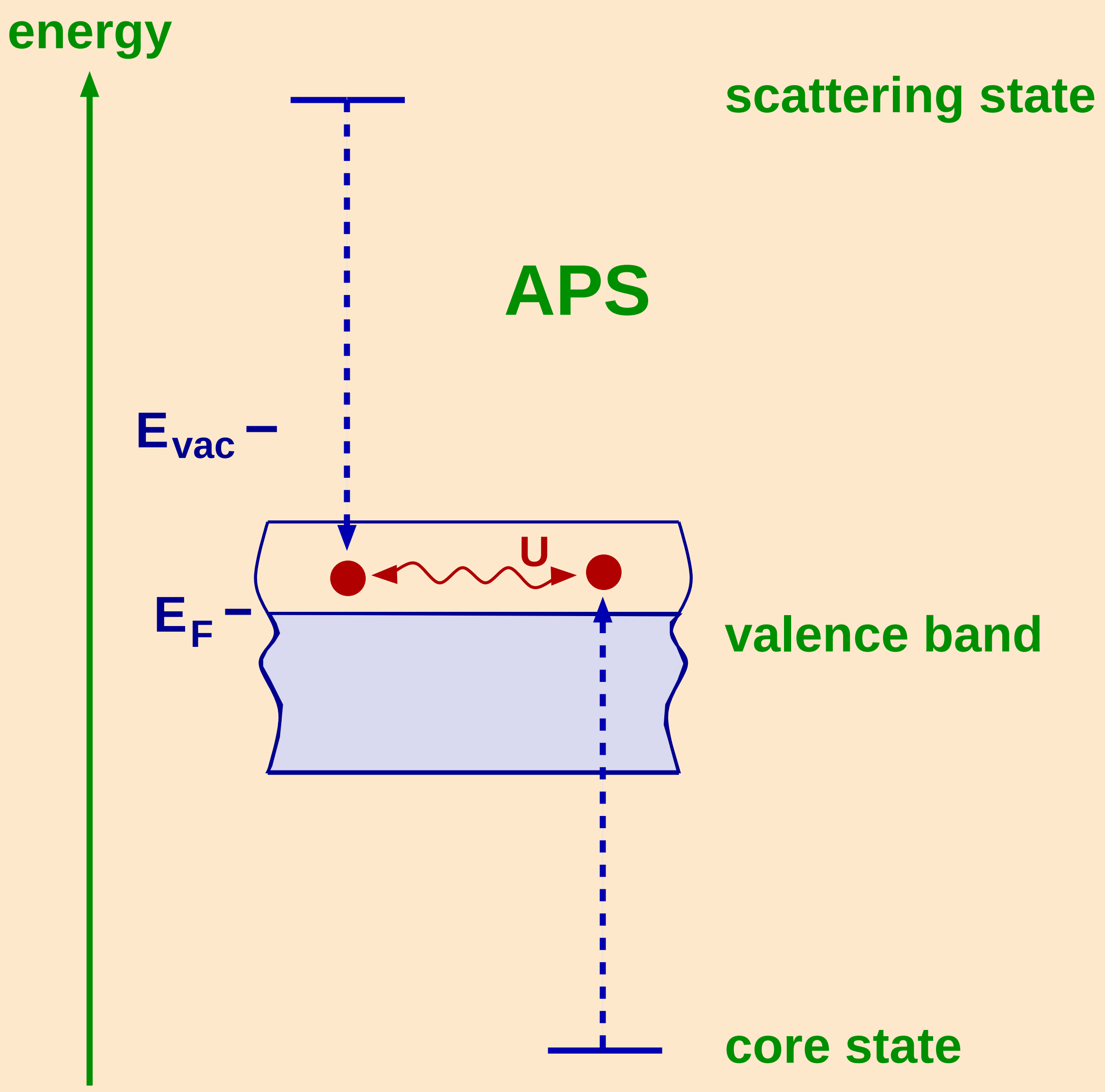
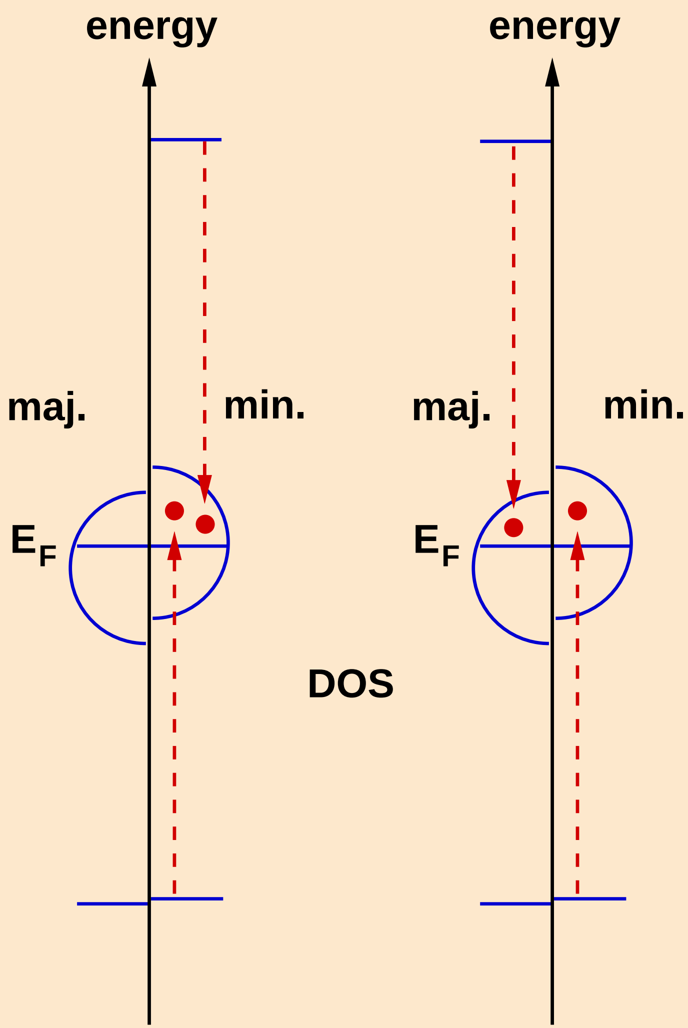

energy
scattering state
APS
E
vac
U
E
F
valence band
core state

outline

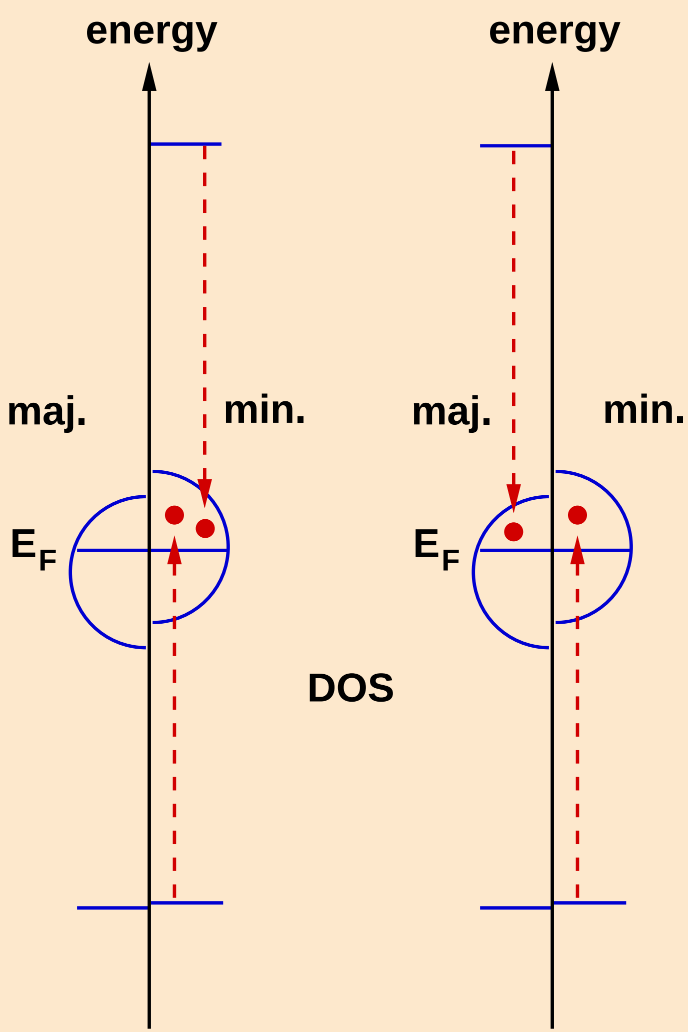
- appearance-potential spectroscopy
- spectrum for ferromagnetic Ni
- interpretation
- different theoretical approaches
- correlation effects ?
- conclusions

experiment



- Ni(110), in-plane magnetization
- polarized electron beam ($P \approx 30\%$)
- core-hole decay detected via X-ray emission (SXAPS)
- lock-in technique, differential spectra

experiment



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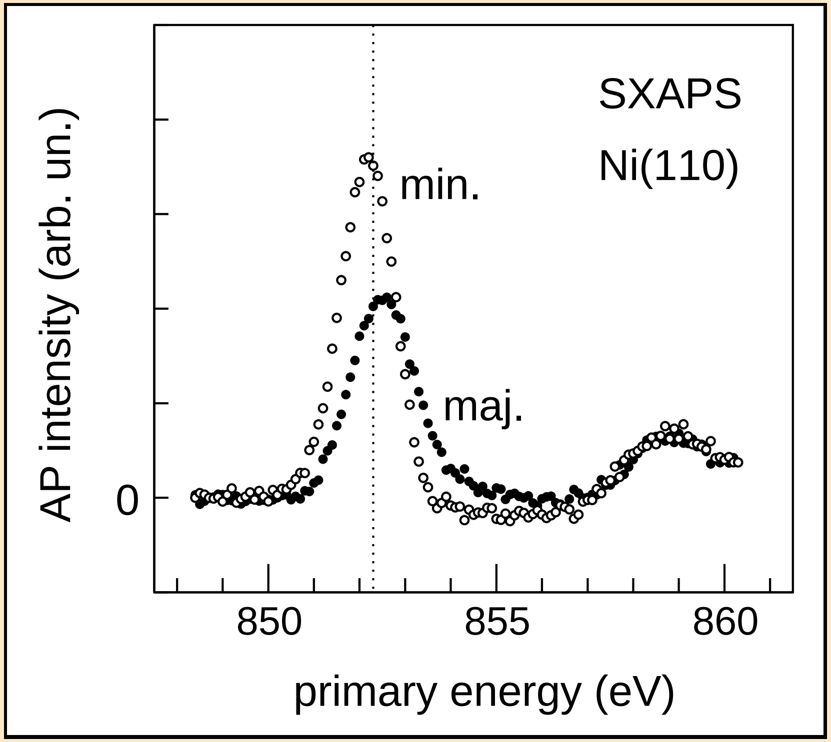
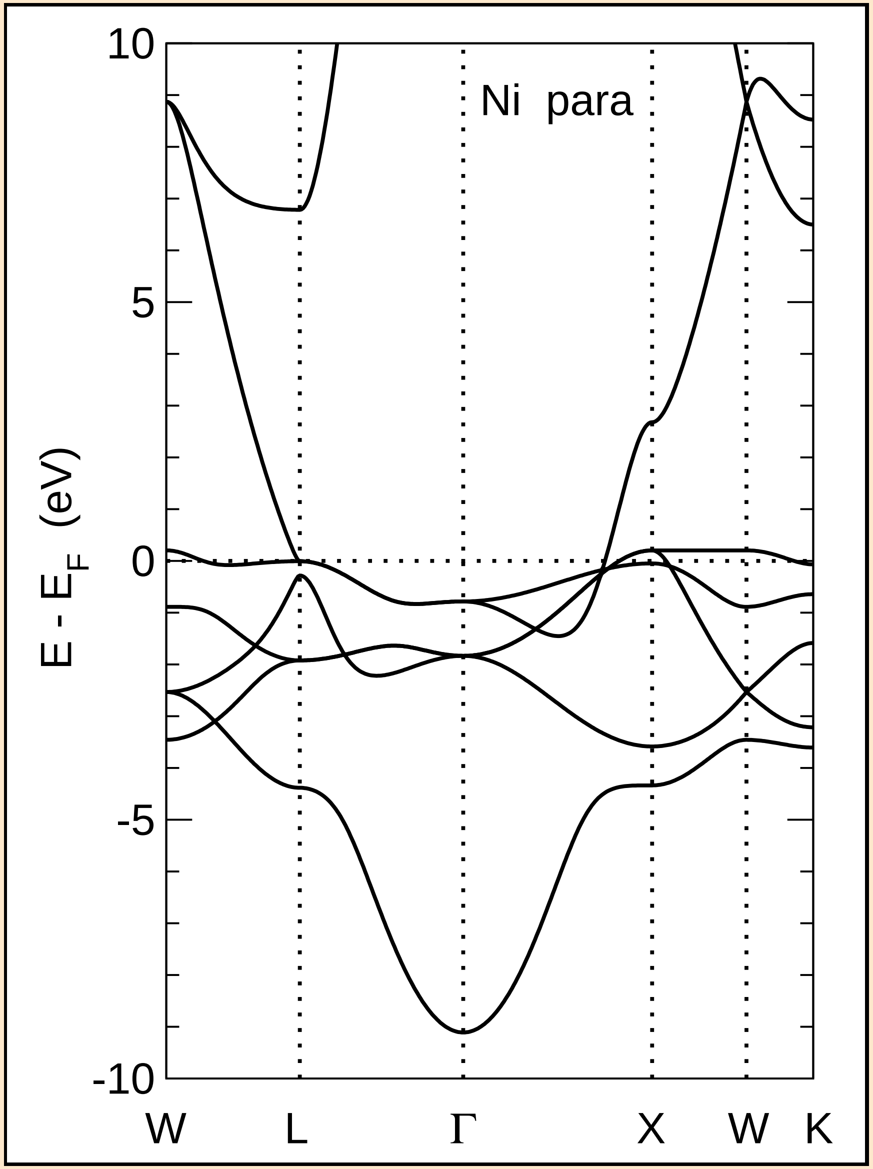


LDA bands



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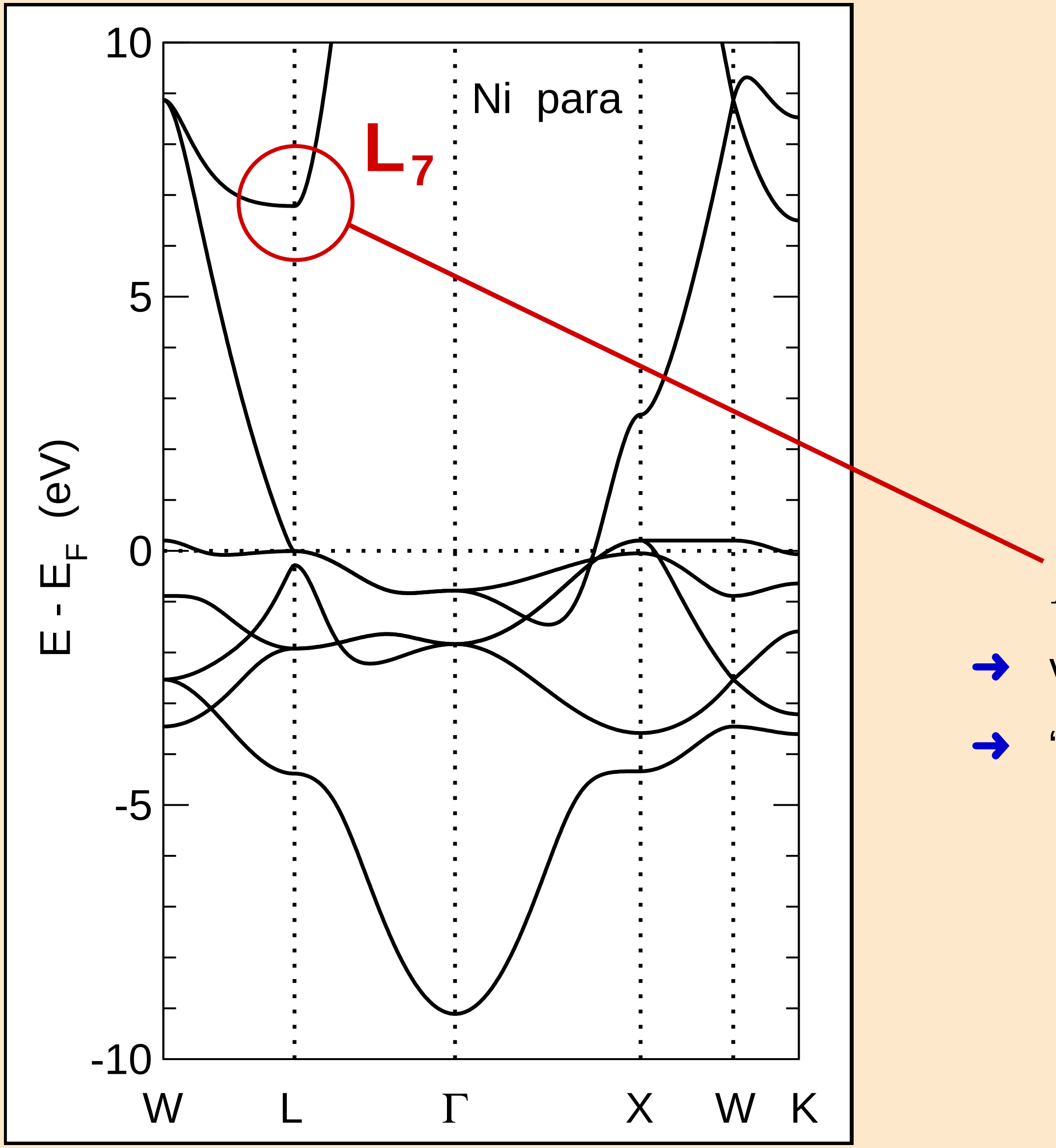
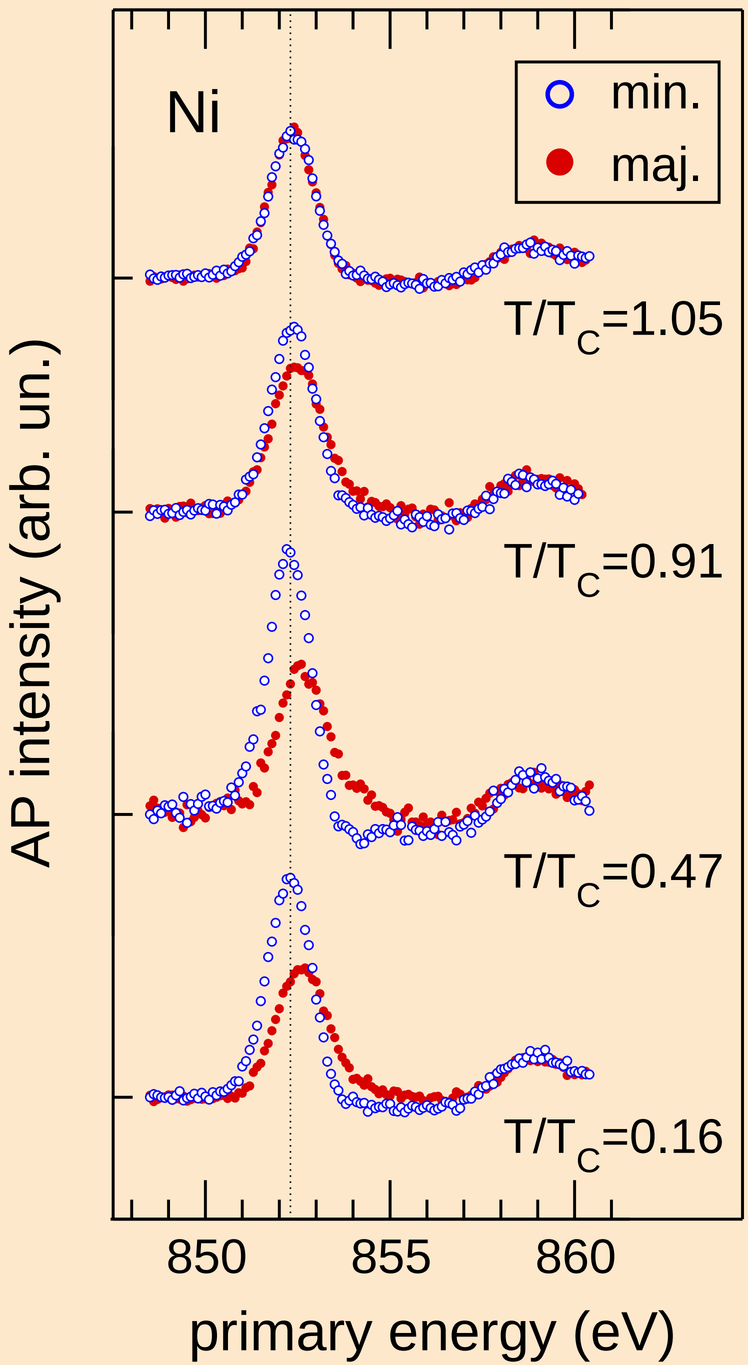


L_7 critical point

➜ van Hove singularity

➜ “structure peak”

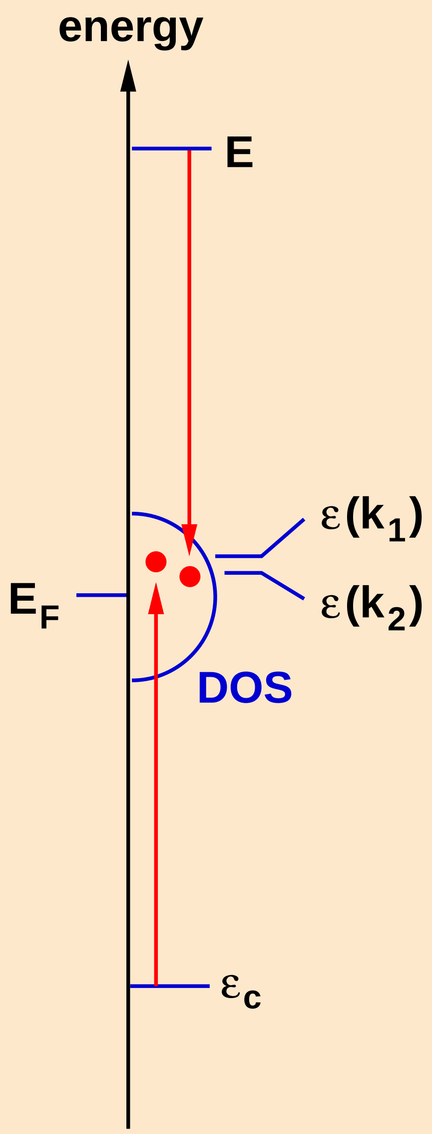
temperature dependence



increasing T up to $T_C \approx 630$ K

- ➜ decreasing spin asymmetry
- ➜ decreasing spin splitting
- ➜ shift of majority peak only
- ➜ structure peak T independent

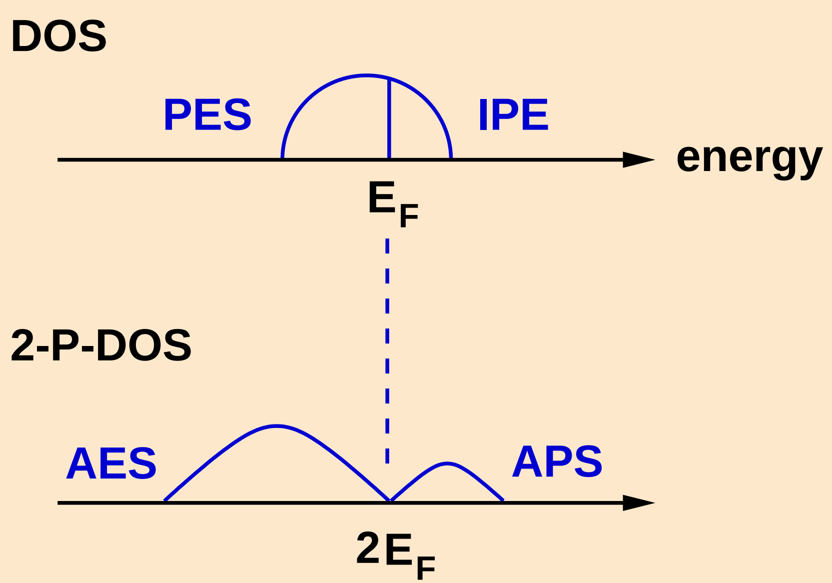
energy conservation



$$E + \epsilon_c = \epsilon(\mathbf{k_1}) + \epsilon(\mathbf{k_2})$$

$$\boxed{I^{\text{APS}}(E) \propto \int dE' \, \rho^{\text{unocc.}}(E') \, \rho^{\text{unocc.}}(E - E')}$$

(Lander's self-convolution model)



basic theory

- Fermi's golden rule
- sudden approximation
- intra-atomic transition

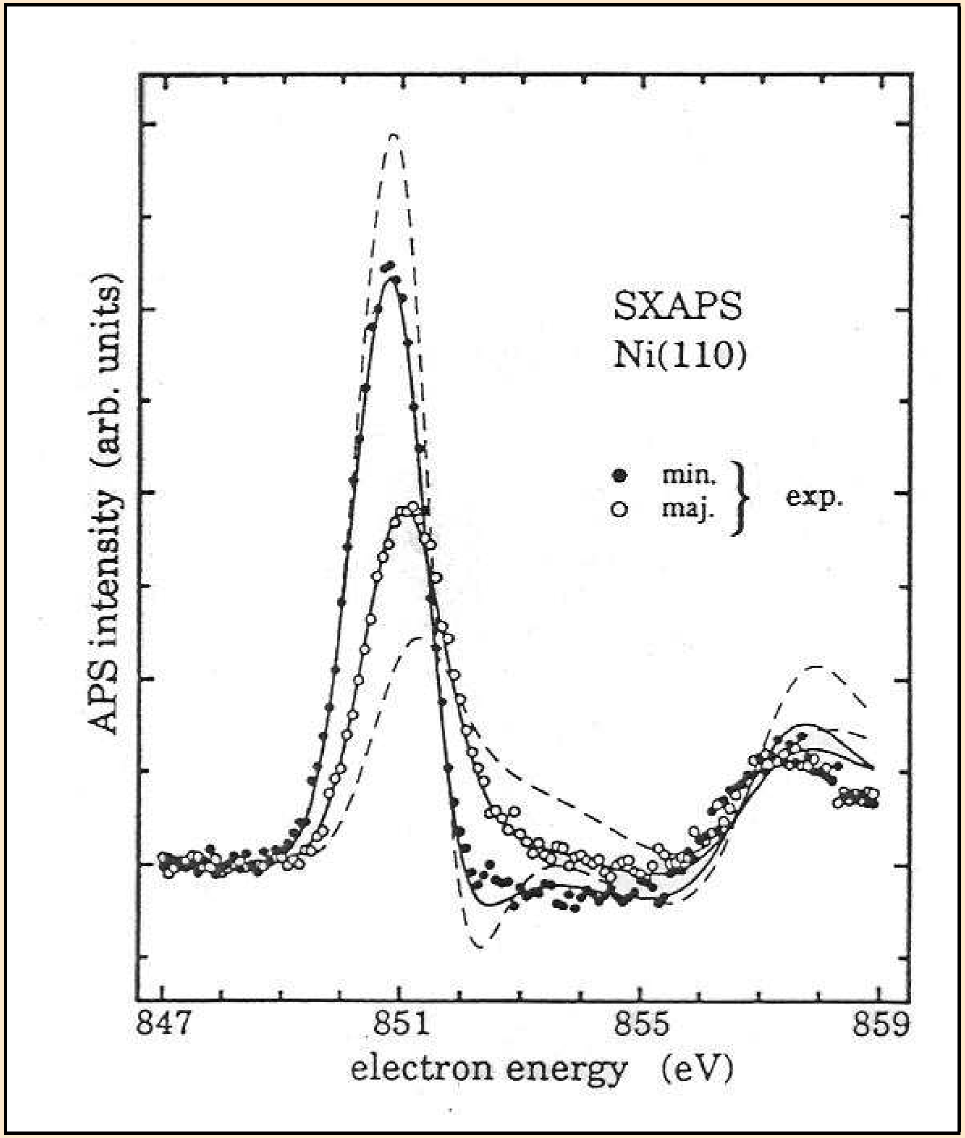
$$I_{\sigma_c \sigma_i}(\mathbf{k}_{\|}, E) \propto \text{Im} \sum_{\substack{L_1 L_2 \\ L_1' L_2'}} M^{\sigma_c \sigma_i}_{L_1 L_2}(\mathbf{k}_{\|}, E) \; \langle\langle c_{iL_1\sigma_c} c_{iL_2\sigma_i} ; c^\dagger_{iL_2'\sigma_i} c^\dagger_{iL_1'\sigma_c} \rangle\rangle_E \; \left(M^{\sigma_c \sigma_i}_{L_1' L_2'}(\mathbf{k}_{\|}, E)\right)^*$$

"raw spectrum": two-particle Green function $\langle\langle c \, c \, ; c^\dagger c^\dagger \rangle\rangle$
transition-matrix elements: $M^{\sigma_c \sigma_i}_{L_1 L_2}(\mathbf{k}_{\|}, E) = \langle 2\text{p}, \sigma_c | \, \langle \mathbf{k}_{\|} E \sigma_i | \, r_{12}^{-1} \, | L_1 \sigma_c \rangle \, | L_2 \sigma_i \rangle$

three steps of improvement compared to Lander's model

- orbital degeneracy
- matrix elements
- correlation effects

orbital degeneracy



Ertl et al. (1993)

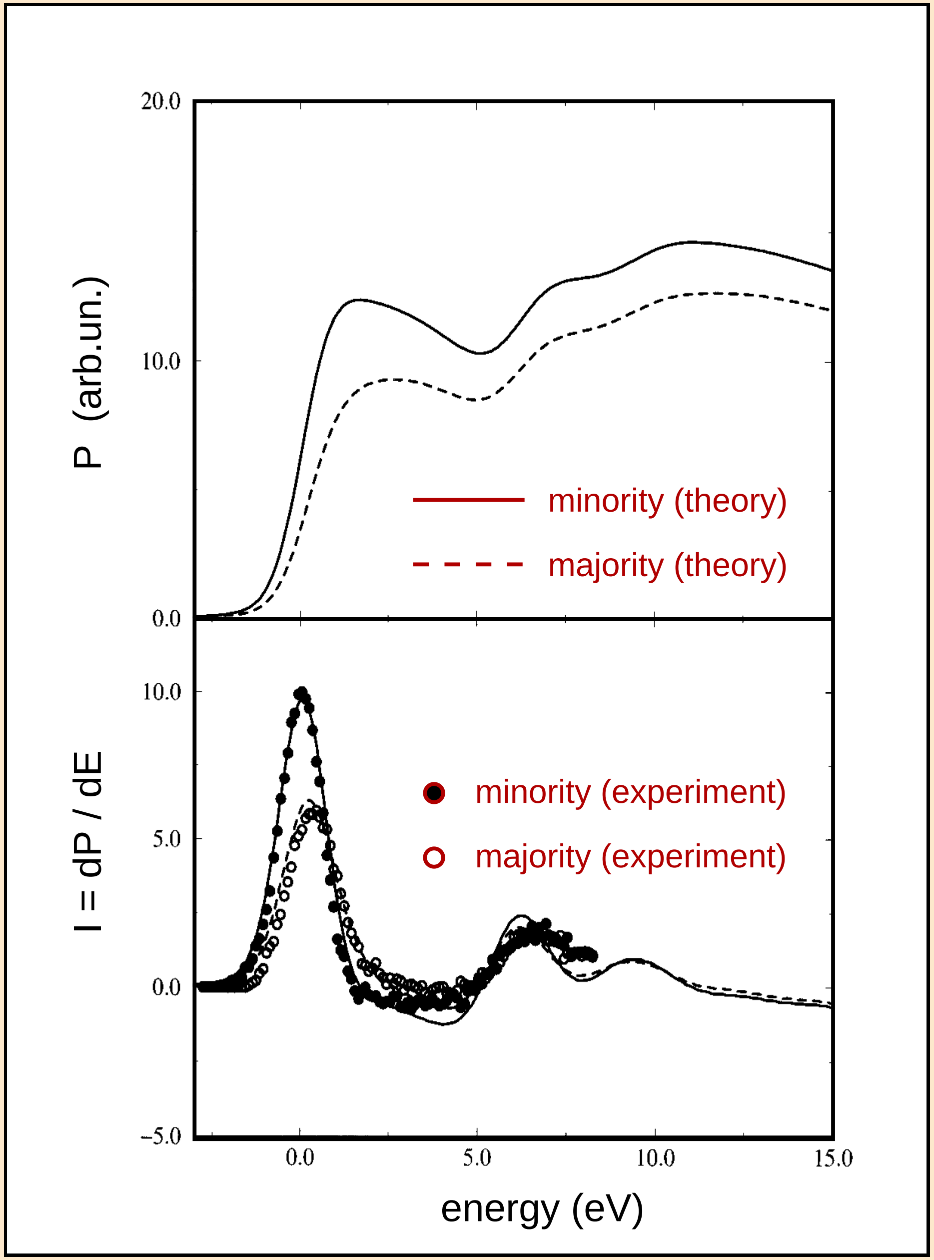
Lander model: - - - - - -
self-convolution of total DOS

s-p-d model: ———
weighted sum of self-convolutions

fit of matrix elements:

$d_\uparrow d_\uparrow$	0.75
$d_\downarrow d_\downarrow$	0.22
$d_\uparrow d_\downarrow$	1.0
$p_\sigma d_{\sigma'}$	0.25
$p_\sigma p_\sigma$	0.7
$p_\sigma p_{\sigma'}$	0.9
$s_\sigma d_{\sigma'}$	0.2
$s_\sigma p_{\sigma'}$	0.0
$s_\sigma p_{\sigma'}$	0.0
$s_\sigma s_{\sigma'}$	0.0

matrix elements



- ab-initio approach based on DFT-LDA
- self-convolutions weighted by matrix elements

→ s-s, p-p, s-p contributions small
→ s-d, p-d contributions important

Ebert and Popescu (1997)

Hubbard-type model

$$H = \sum_{ii'LL'\sigma} t_{ii'}^{LL'} \, c_{iL\sigma}^\dagger c_{i'L'\sigma} + \frac{1}{2} \sum_{i\sigma\sigma'} \sum_{L_1 \dots L_4} U_{L_1 L_2 L_4 L_3} \, c_{iL_1\sigma}^\dagger c_{iL_2\sigma'}^\dagger c_{iL_3\sigma'} c_{iL_4\sigma}$$

- 3d, 4s, 4p states (9 orbitals per site)
- (non-orthogonal) MTO's $|iL\sigma\rangle$
- full (on-site) Coulomb matrix
- $U = 2.47$ eV, $J = 0.5$ eV

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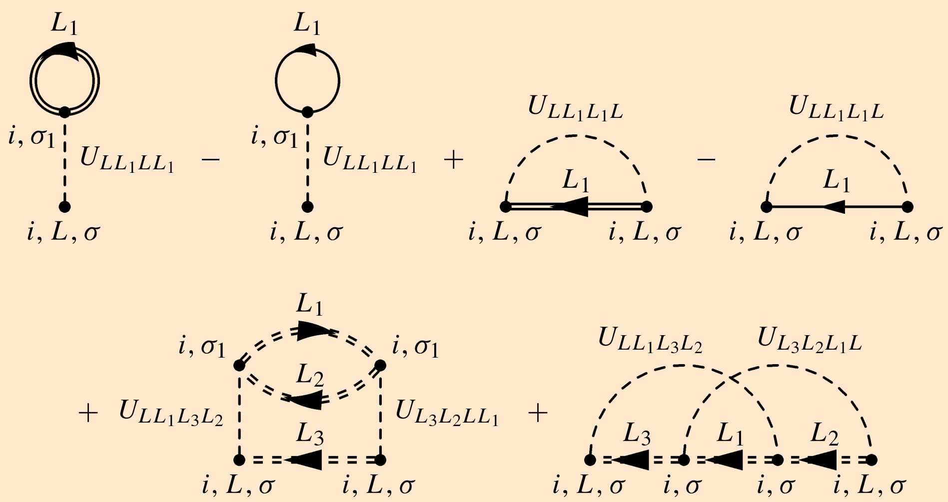
SOPT−HF self−energy



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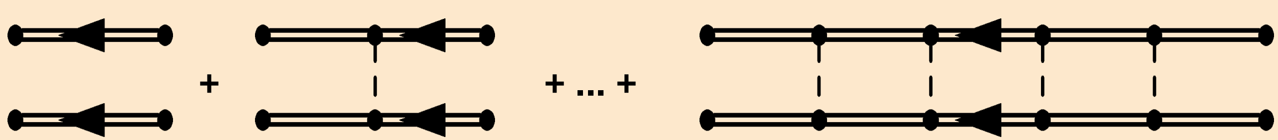
ladder approximation



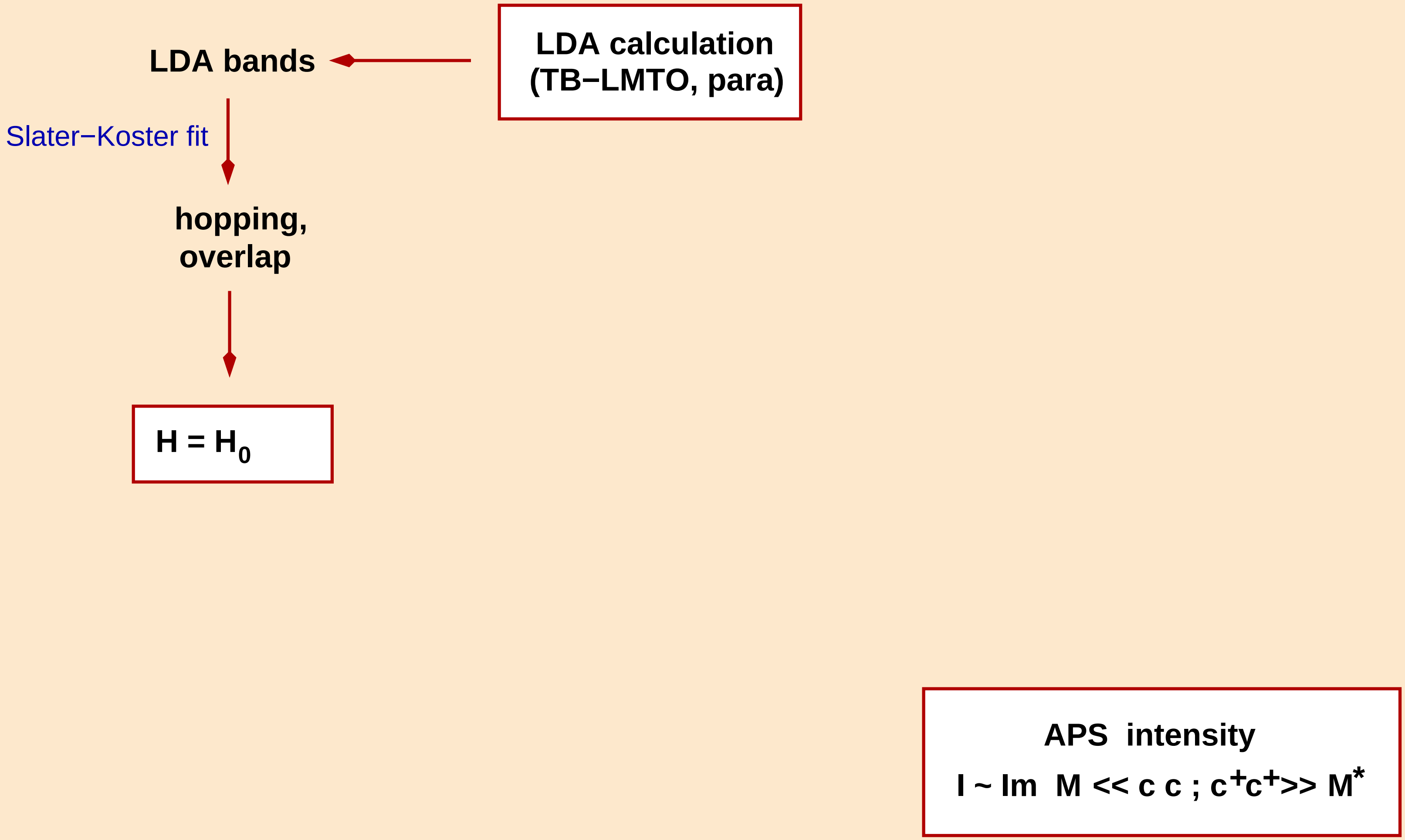
calculations

LDA calculation
(TB−LMTO, para)

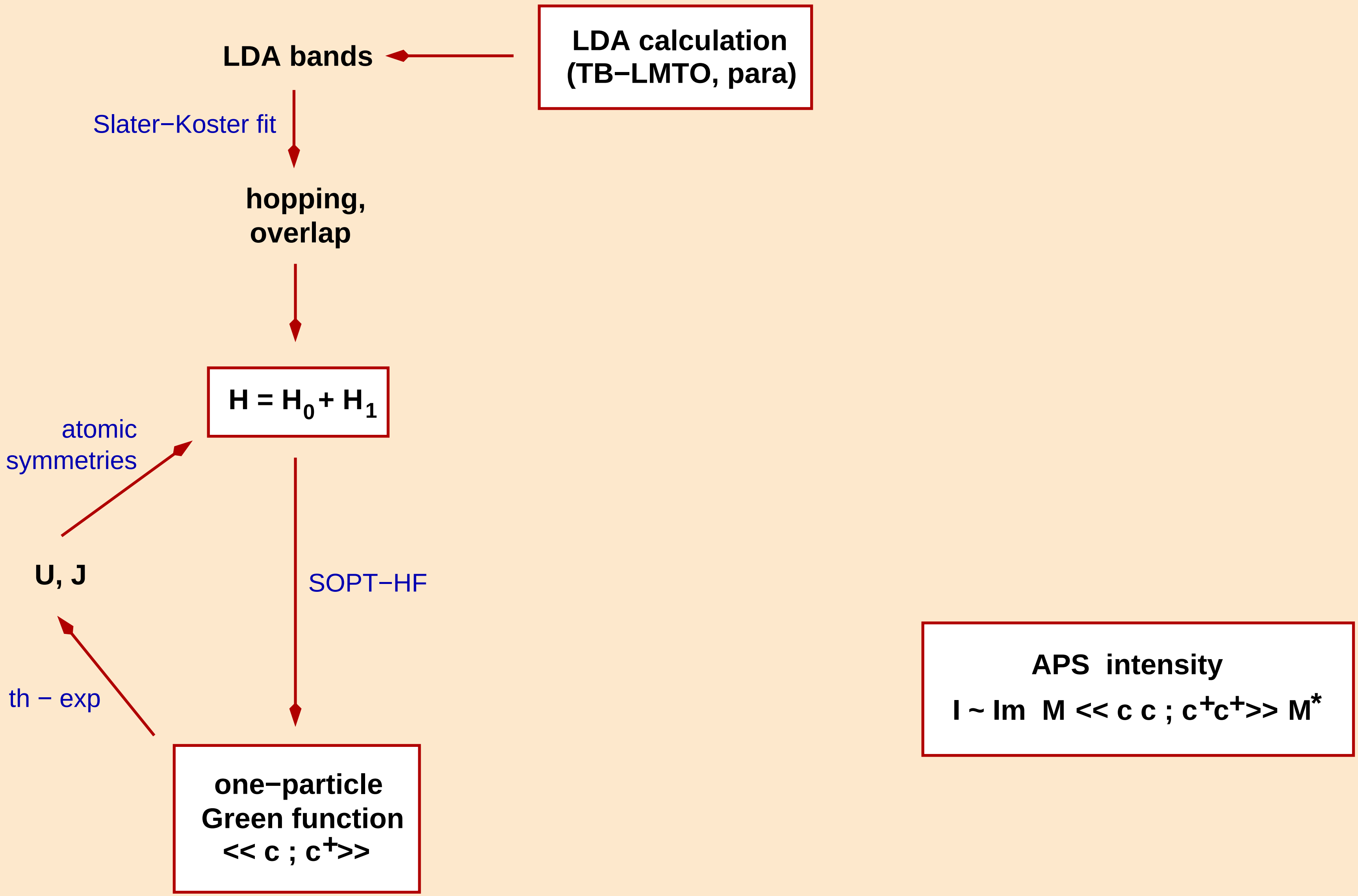
APS intensity

$I \sim \mathrm{Im}\ M \langle\langle c\, c\, ; c^{+} c^{+} \rangle\rangle M^{*}$

calculations



calculations



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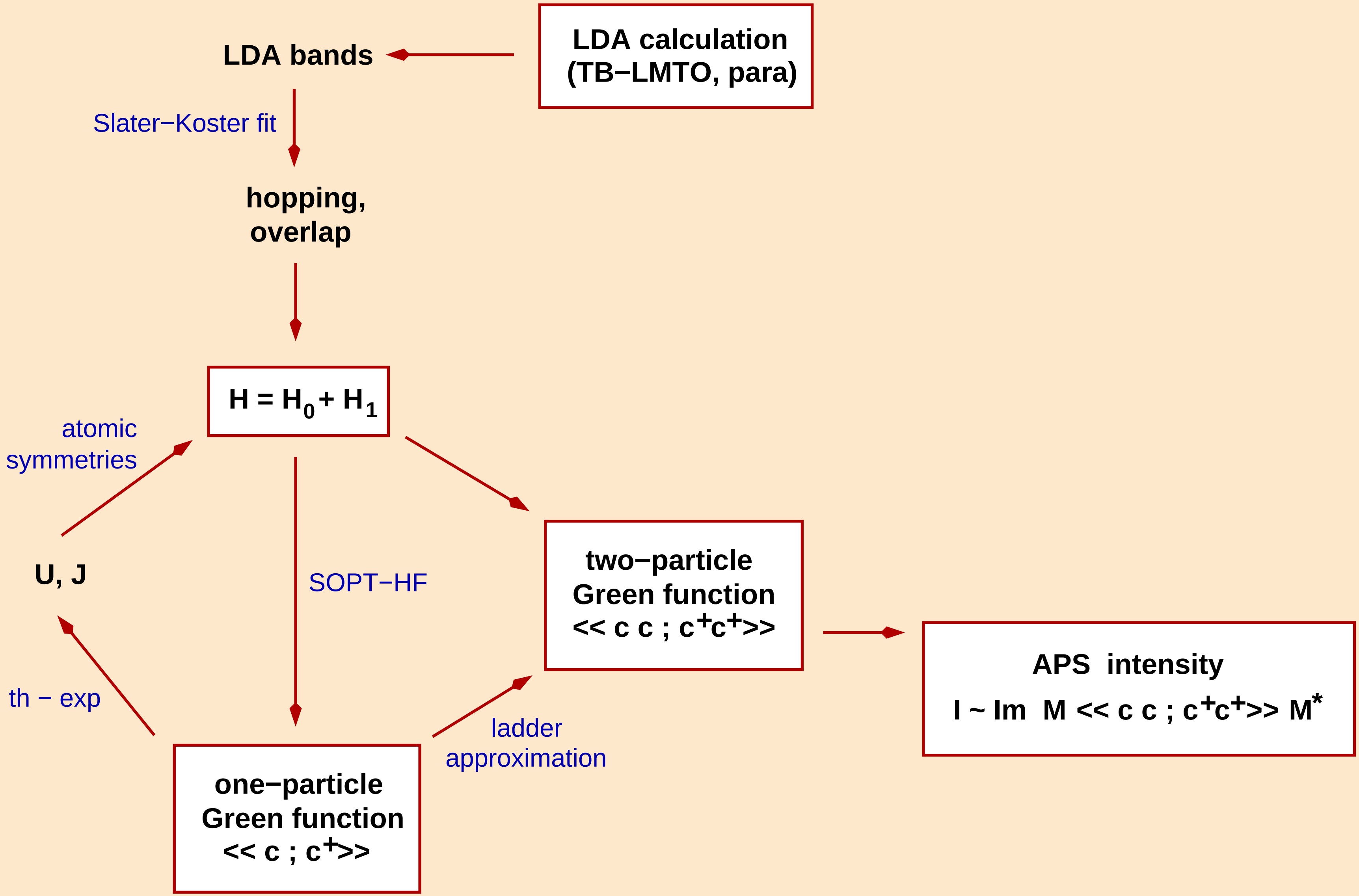
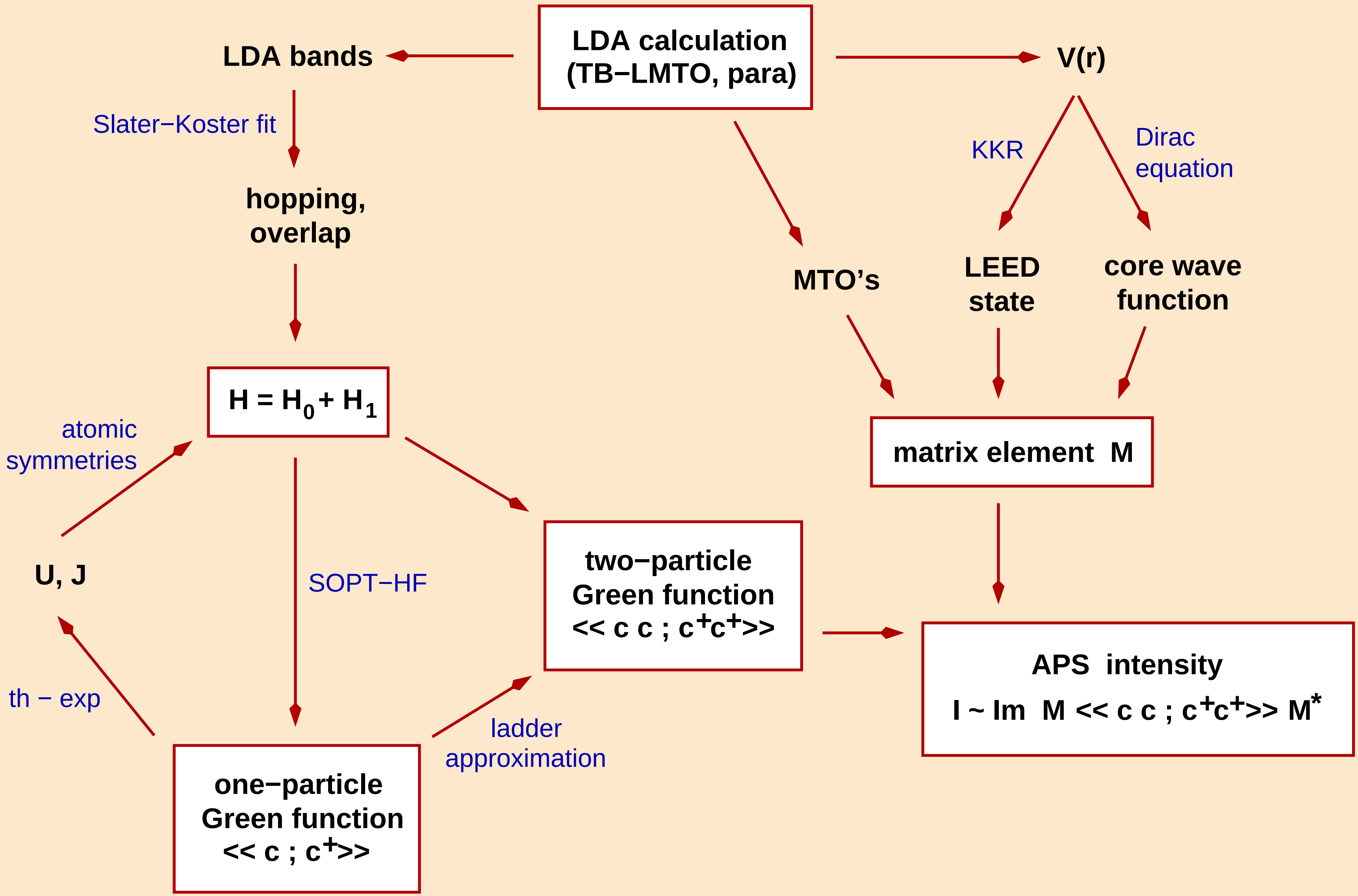
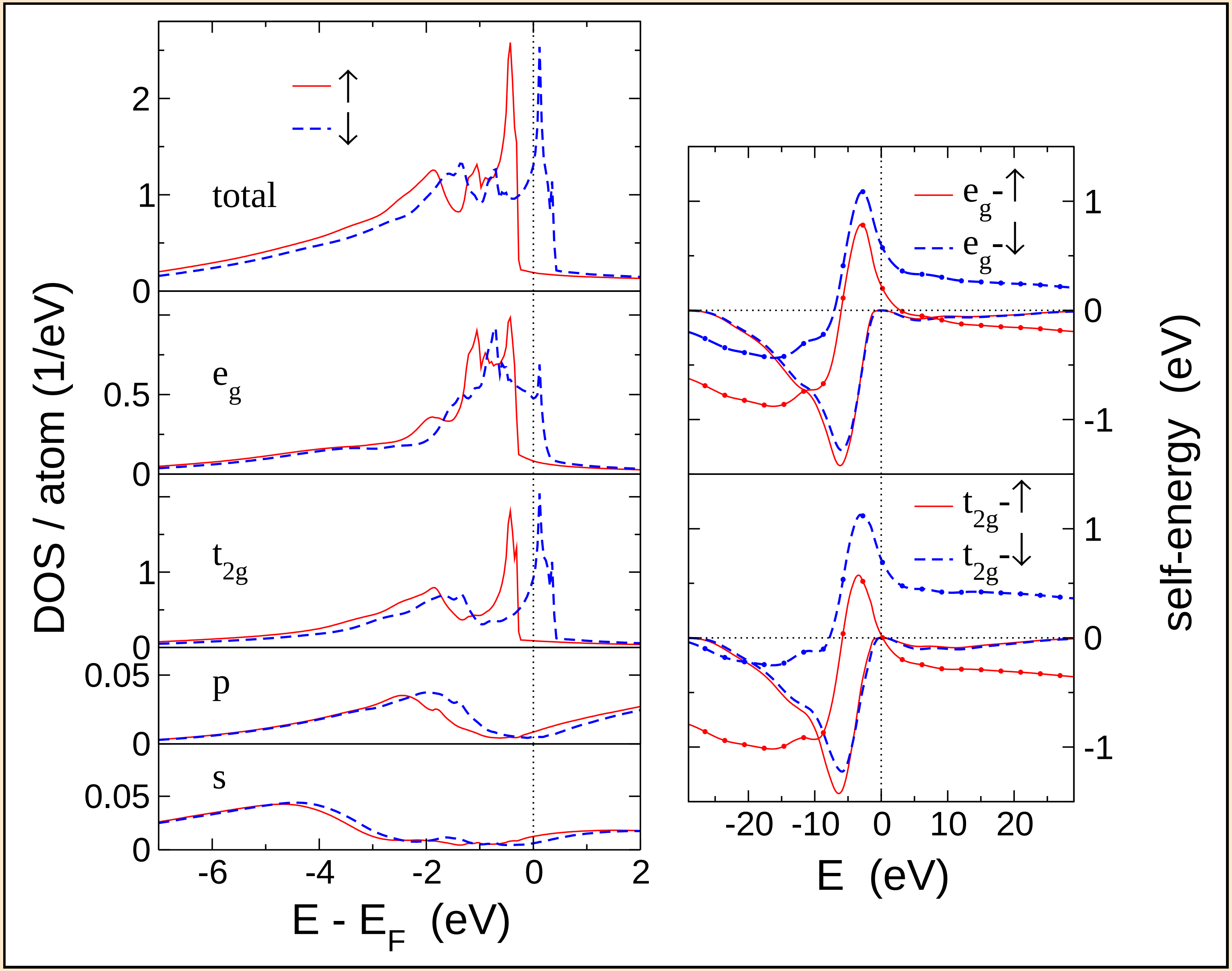


calculations

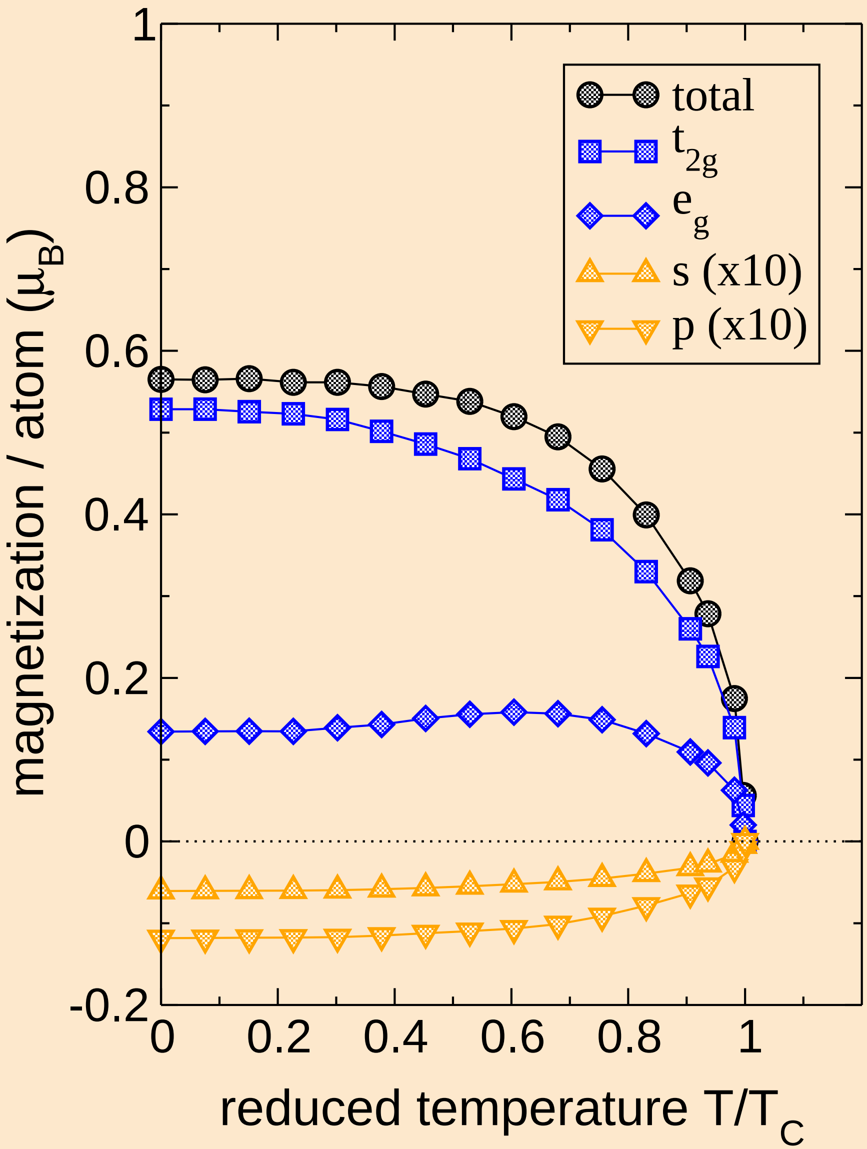


SOPT-HF results



- band narrowing
- life-time effects

Ni ferromagnetism

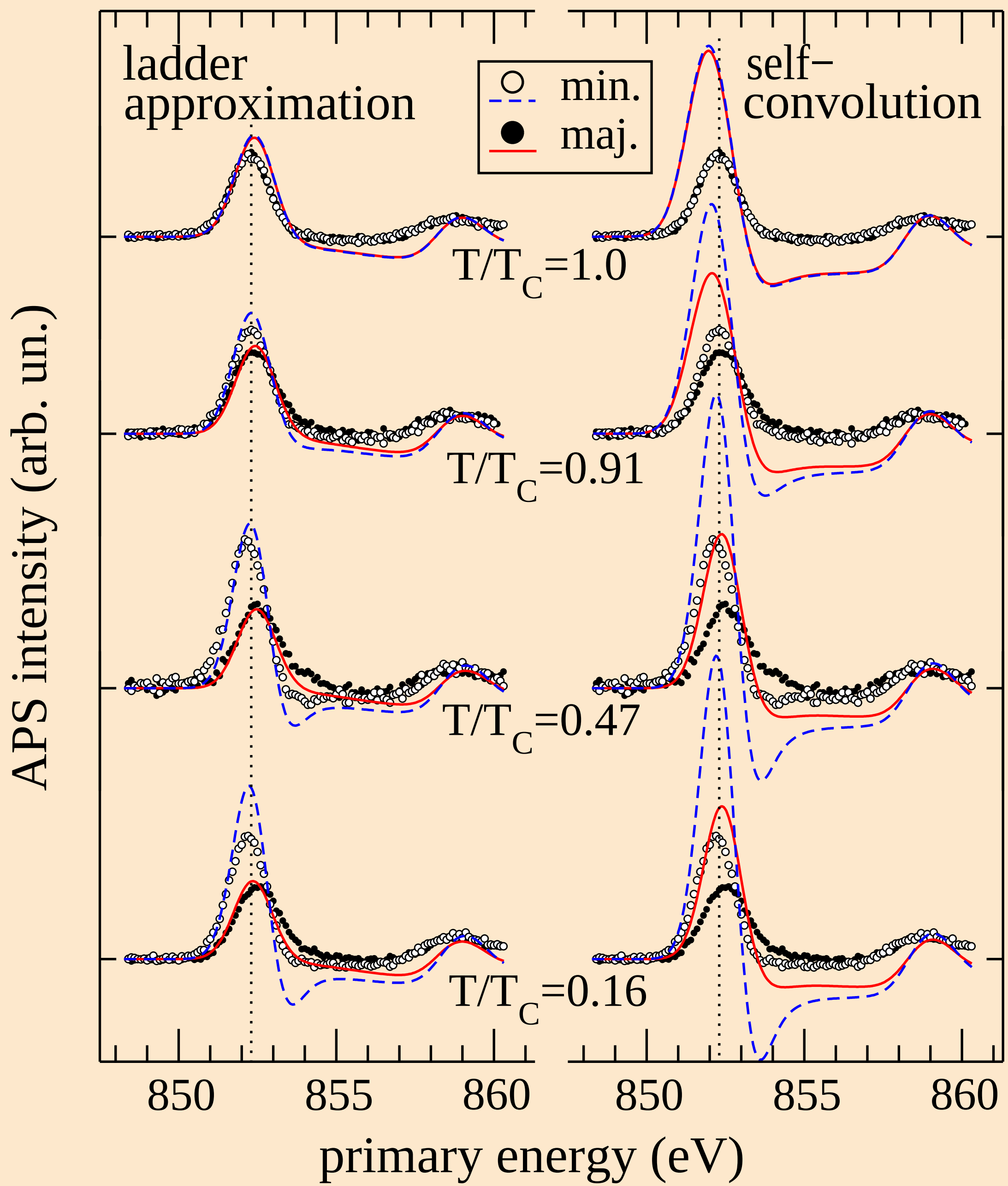


Curie temperature

- ➜ measured: $T_C \approx 630$ K
- ➜ calculated: $T_C = 1655$ K

- $T = 0$ magnetic moment
- temperature trend of m
- antiferromagnetic alignment between d and s-p moment

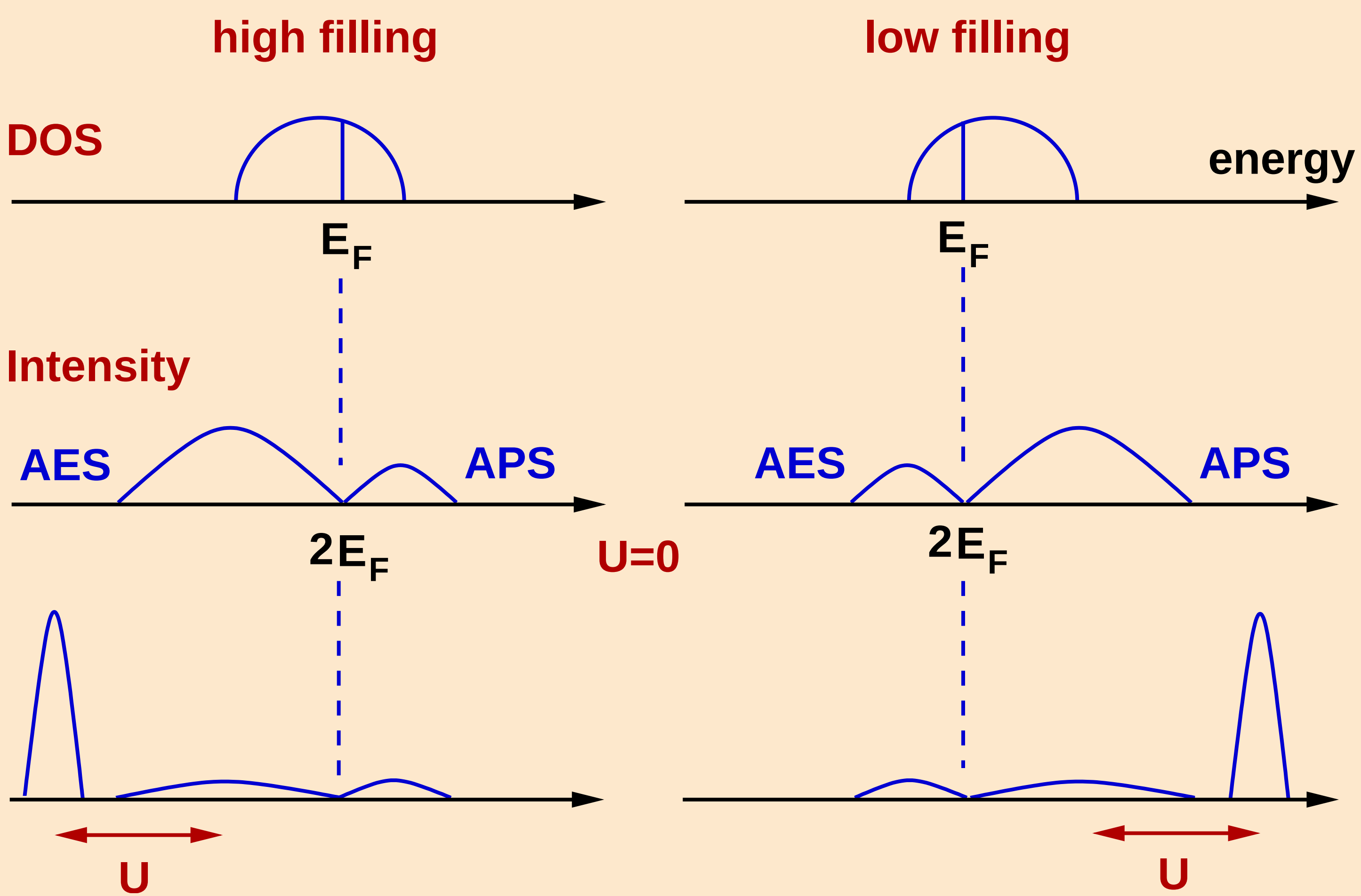
APS spectrum



M. Potthoff, T. Wegner, W. Nolting, T. Schlathölter, M. Vonbank, K. Ertl, J. Braun, and M. Donath (2001)

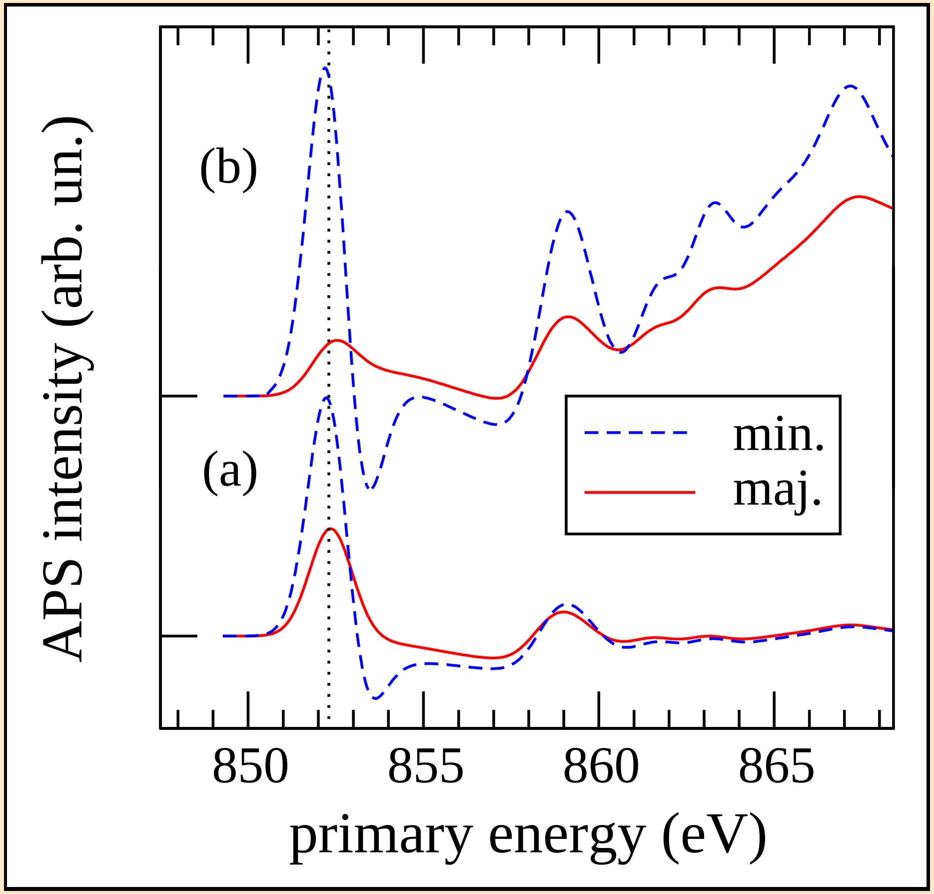
Cini-Sawatzky theory

- single-band Hubbard model
- on-site Coulomb interaction U
- $I \propto \langle\langle c\, c\, ; c^\dagger c^\dagger \rangle\rangle$
- exact calculation for $n = 0$, $n = 2$
- extrapolated by ladder approximation



matrix elements

$$M^{\sigma_c \sigma_i}_{L_1 L_2}(\mathbf{k}_\parallel, E) = \langle \text{2p}, \sigma_c | \, \langle \mathbf{k}_\parallel E \sigma_i | \, r_{12}^{-1} \, | L_1 \sigma_c \rangle \, | L_2 \sigma_i \rangle$$

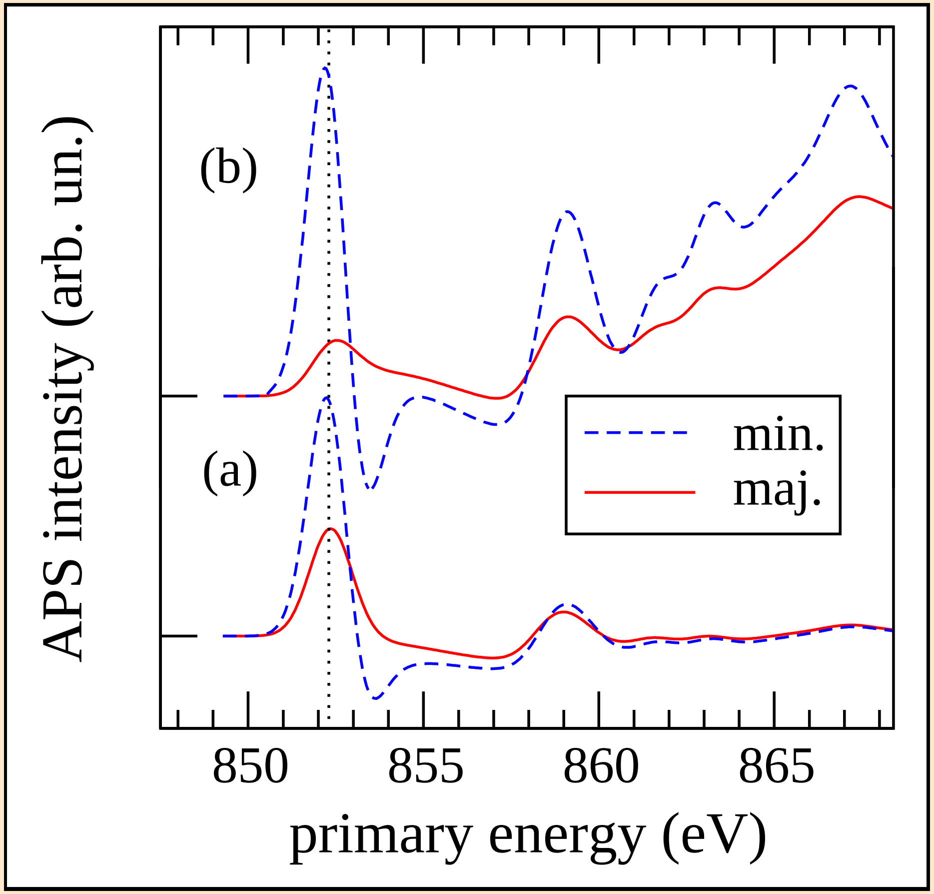


(a) full theory

(b) matrix elements set constant: $M^{\sigma_c \sigma_i}_{L_1 L_2}(\mathbf{k}_\parallel, E) \mapsto M_{L_1 L_2}$ = const

matrix elements

$$M^{\sigma_c \sigma_i}_{L_1 L_2}(\mathbf{k}_{\|}, E) = \langle 2\text{p}, \sigma_c | \langle \mathbf{k}_{\|} E \sigma_i | \, r_{12}^{-1} \, | L_1 \sigma_c \rangle \, | L_2 \sigma_i \rangle$$



➜ suppression of s-p contributions

➜ control of spin asymmetry:

$M_{L_2 L_2} = +M_{L_1 L_2}$ ➜ $I_\uparrow = I_\downarrow$

$M_{L_2 L_2} = -M_{L_1 L_2}$ ➜ maximum spin asymmetry

(a) full theory

(b) matrix elements set constant: $M^{\sigma_c \sigma_i}_{L_1 L_2}(\mathbf{k}_{\|}, E) \mapsto M_{L_1 L_2} =$ const

conclusions

❖ **experiment:**

– **spin-resolved and temperature-dependent APS of Ni**

❖ **theory:**

– **orbital degeneracy**

– **transition-matrix elements**

– **correlation effects (perturbational)**

➜ **Lander model insufficient**

➜ **spin asymmetry controlled by matrix elements**

➜ **correlation-induced spectral weight transfer**

➜ **quantitative agreement with experiment using different theories — error cancellations !?**

? **core-hole effects in the final state**